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# Portable NDA Equipment for Enrichment Measurements in the HEU Transparency Program

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### Abstract

The Highly Enriched Uranium (HEU) Transparency Program has used portable non-destructive assay (NDA) equipment to measure the <sup>235</sup>U enrichment of material subject to the transparency agreement since 1997. The equipment is based on the "enrichment meter" method and uses low-resolution sodium iodide (NaI(Tl)) detectors. Although systems using high-purity germanium (HPGe) detectors can produce more accurate results we have found that the results with NaI(Tl) detectors are quite adequate for the requirements of the transparency agreement. This paper will describe the details of the equipment's operation, calibration, testing, and deployment in Russia. We will also provide a comparison of the units originally deployed in 1997 with the upgraded systems that were deployed in 2003.

### Introduction

In 1996 the US and Russia agreed to use non-destructive assay (NDA) equipment for uranium enrichment measurements for the HEU Transparency Program. The systems were to be composed of commercially available components and needed to be able to measure the <sup>235</sup>U enrichment of uranium in a variety of container types and chemical forms. Furthermore the system needed to be able to measure freshly processed uranium. These requirements led to the selection of systems based on the uranium enrichment meter principle. The systems were based on low-resolution sodium iodide detectors which allowed a compact, low-cost and low-maintenance alternative to HPGe detectors.

The enrichment meter method has been used since the 1970's and is the standard technique for uranium enrichment measurements for the International Atomic Energy Agency (IAEA). This topic is described in detail in reference 1. In this method, the enrichment of the sample is proportional to the rate of the 186-keV gamma rays emitted by <sup>235</sup>U. The method is valid for measuring the enrichment of bulk uranium samples that are homogeneous and large enough to fill the field of view of the gamma-ray detector with a so-called infinite thickness of uranium.

The infinite thickness is defined as that amount of material that can attenuate the intensity of the 186-keV gamma ray by a factor of one hundred. For uranium metal this thickness is about 3 millimeters; for solid UF<sub>6</sub> it is 1.4 centimeters. An appropriate collimator restricts the field of view of the detector so that reasonably sized samples can fulfill the

requirements of the enrichment meter principle. When these conditions are met we can determine the enrichment by the equation:

$$Enrichment = K \cdot R(186) \cdot C_{Mat} \cdot e^{-\mu t}$$

where R(186) is the observed net peak count rate of the 186 keV gamma ray in the detector,  $C_{Mat}$  is a correction for the type of uranium material, and the exponential factor corrects for the attenuation of the gamma rays by the container wall. The term  $C_{Mat}$  is quite small, if we normalize such that the  $C_{Mat}(U_30_8) = 1.00$  then the correction for uranium metal is 0.98 and that for UF<sub>6</sub> is 1.022. The calibration constant K must be determined with a suitable uranium source of known enrichment.

The analysis of the data then consists of extracting the "peak area" of the 186-keV gamma-ray line from the NaI(Tl) spectrum of the HEU sample. Because of the low energy resolution of NaI(Tl) detectors this process can produce varying results since there are so few channels available for estimating the Compton continuum. This is particularly true for the low-energy side of the 186-keV peak. In order to minimize the fluctuation associated with the delineation of the background one can use the "two-window" approach to the enrichment meter method. Here we define the enrichment in terms of the gross counts in a window about the 186-keV line and a second background window at higher energy. In this way the enrichment is expressed as:

$$Enrichment = (AR_1 + BR_2) \cdot C_{Mat} \cdot e^{-\mu t}$$

In this equation  $R_1$  and  $R_2$  are the measured peak and background count rates, respectively. The constants "A" and "B" are determined by calibrating the system with two standards of different enrichments. This method is the standard algorithm used by the IAEA in its procedures to measure uranium enrichment with NaI(Tl) detectors.

The systems used by the HEU Transparency Program are composed of a collimated NaI(Tl) scintillation detector, a portable multichannel analyzer (MCA) and a laptop computer. The detector is a 1 inch by 1 inch NaI(Tl) crystal coupled to a photo multiplier tube. The detector's field of view is restricted by a collimator with a length of 0.5 inches and an aperture of 0.5 inches. In addition, a lead shield runs the length of the detector, thus reducing the background count rate and shielding the detector from other uranium sources in close proximity. We calibrate the systems with uranium standards at the Lawrence Livermore National Laboratory or at the Y-12 plant in Oak Ridge, TN.

These systems are described in more detail in reference 2. This equipment performed well but had more capability than was really necessary for these measurements. Also these early systems had somewhat complicated cable connections. After five years of use in the Russian facilities the program changed to a different type of equipment. These units which were deployed in 2003 and 2004 are described below.

# Description of currently used equipment

The new units are based on a more compact MCA with a matching sodium iodide detector. The MCA is small enough that it could be mounted directly on the collimator assembly. Also there is only one cable between the detector and MCA and this cable can be left attached during storage. This greatly reduces the failure rate of the cable and connectors. The MCA can be small since it does not contain the high voltage (HV) supply or amplifier for the detector; both of these components are built into the tube base of the SCIONIX detector. Figure 1 shows a picture of one of the units.



Figure 1: Photograph of the portable NDA equipment used by the HEU Transparency Program

The multichannel analyzer – NaI(Tl) detector system, designated by the trade name "GAMMA-8000" was purchased from AMPTEK, Inc. The unit is composed of the AMPTEK MCA8000A "pocket MCA" and the specially adapted, low-power, NaI(Tl) detector from SCIONIX. Other components of the complete NDA system include the collimator assembly, a laptop computer, the RS-232 communication cable and AA sized batteries. The detector has a 30 mm by 30 mm NaI(Tl) crystal, thick enough for high peak efficiency detection of the 186-keV gamma rays, and the surface area is more than adequate for the high count rates for HEU measurements. Figure 2 shows a picture of the components.



Figure 2: Photograph of the components for the portable NDA equipment.

The vendor of the MCA supplied a data acquisition, display, and analysis code called PMCA. This code is useful for testing the detector and MCA but is not adequate for our application. However, the vendor also supplies a programming library with routines for communicating with the MCA. We used these routines to develop a software package, written in Visual Basic, for the enrichment measurements. We have successfully operated the software based on these routines with computers running Windows 95, 98, 2000 and XP. A more detailed discussion of the use of the software and hardware is given in reference 3.

The original equipment that was based on the Canberra Inspector used a software package that was written as a REXX procedure in OS/2. This menu-driven procedure worked well and had sufficient flexibility for this application. The software for the AMPTEK based device, called UM\_2001, was written to emulate this earlier code. The UM\_2001 program flow, shown in Figure 3, is quite simple. The code first establishes communication with the MCA and powers it on. Once this is accomplished the user is presented with a menu consisting of several pre-set container types to measure, a user-specified container option and an exit function. The program then allows the operator to enter the data acquisition time, usually 60 seconds, and then begins the data acquisition. The progress of the data acquisition is displayed by a countdown timer and by showing the changes in the pulse height spectrum. The spectral display allows the operator to check that the instrument is performing properly, if it is not, the operator can abort the run to make adjustments. When the data have been acquired the program immediately

calculates and displays the enrichment if the operator had selected one of the pre-set container options. For the "User-specified" option the operator must enter the material type (metal, oxide, or hexafluoride), the container wall material, and the container wall thickness. Once these parameters are entered the program will then calculate and display the enrichment value. After the operator acknowledges the result by clicking the "OK" button in the results window the program returns the user to the original menu for more measurements or to exit the code.

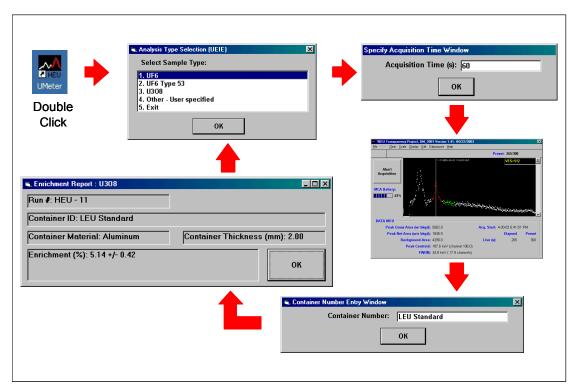


Figure 3: Flow diagram of the computer code for uranium enrichment measurements.

One key feature of the code is its ability to mitigate the effects of gain shifts and drifts in the NaI(Tl) detector spectrum. Hardware gain stabilization is not available with these units. Also there is no software control for the gain since the amplifier and HV supply are set in the tube base. The only fine gain adjust that the user has is a twenty-turn potentiometer on the tube base of the SCIONIX detector. The UM\_2001 program gets around these limitations by using a "sliding-window" analysis. This method changes the position of the analysis windows (the determination in R1 and R2 in the second equation) to account for small changes in the position of the 186-keV gamma ray peak in the spectrum. The code displays error messages if it needs to make a large shift (> 10 keV) in these windows. The operators are trained to make an adjustment of the potentiometer when these error messages appear. This software feature enables the systems to be reliably used from one monitoring visit to the next with minimal adjustments.

# Discussion of system performance

We performed extensive testing of the AMPTEK-based systems prior to their deployment. This testing included stability of the energy resolution of the spectrometer, stability of repeated enrichment measurements and the effects of low battery power. The results of these tests showed that the spectral quality of the spectrometer system was sufficient for the needs of this transparency regime. The full width at half maximum for the 186-keV line was about 18 keV and the efficiency of the system is determined by the collimator design rather than the intrinsic efficiency of the NaI(Tl) crystal. A sample spectrum of a US HEU standard is shown in Figure 4. We also determined that two fresh AA batteries would provide sufficient power to perform NDA for an entire typical monitoring visit.

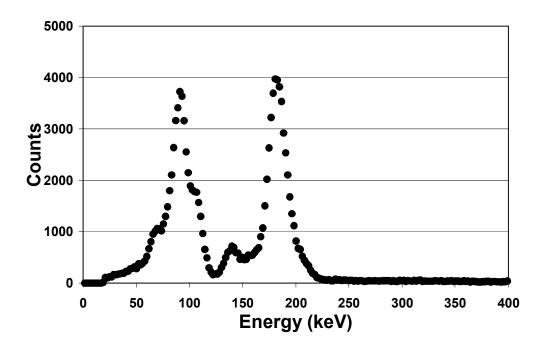


Figure 4: Spectrum of 93% enriched US uranium oxide standard acquired by the portable NDA equipment. The count time was 300 seconds.

The development and testing of these units did point out some areas of the programming that require special attention. These units and the software libraries that support them are based on an asynchronous communication link. The user developed software must provide sufficient checks to ensure that the MCA has actually received and executed commands sent from the computer. There is also a concern about the speed at which commands can be received by the processor in the MCA. We found that as we switched to newer and faster computers the MCA would have difficulty keeping up with a stream of commands. This issue was alleviated by building suitable delay times into the code.

### **Conclusions**

The AMPTEK MCA-based portable NDA units deployed in Russia have performed adequately to meet the needs of this transparency regime. The accuracy of the data for thin-walled containers (3-4 mm) is a few percent, i.e., the expected value from the counting statistics of the one-minute measurements. The units require very little maintenance other than installing new batteries for each visit and an occasional adjustment of the detector gain. The success of the measurements also is due to the skill of the Russian operators and the cooperation of the Russian plant workers and management.

A total of twelve of the NDA systems have been deployed at the four Russian sites that process material for the HEU Transparency Program. In the five years of operation there have been no significant hardware failures and the units have been available to make measurements for all of the monitoring visits. Because the monitoring regime only requires the systems to be used for about ~100 hours per year we expect these units will be available through the scheduled end of the program in 2013.

### References

- 1. Doug Reilly, Norbert Ensslin, Hastings Smith, Jr., and S. Kreiner, "Passive Nondestructive Assay of Nuclear Materials," NUREG/CR-5550, March 1991.
- 2. Decman, D.J, Glaser, J., Hernandez, J.M. and Luke, S.J. (1999) "Portable NDA Equipment for Enrichment Measurements for the HEU Transparency Program". INMM Annual Meeting. Phoenix, Arizona.
- 3. Bandong, B.B., Wong, J.L., Valentine, J.D., Decman, D.J., "Operations Manual for Portable NDA II Equipment", UCRL-MA-149450 (2002).

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